Ann. Geophysicae 12, 365–384 (1994) © EGS – Springer-Verlag 1994

Annales Geophysicae

On the effect of emissions from aircraft engines on the state of the atmosphere

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Received: 28 June 1993/Revised: 24 January 1994/Accepted: 27 January 1994

Abstract. Emissions from aircraft engines include carbon dioxide, water vapour, nitrogen oxides, sulphur components and various other gases and particles. Such emissions from high-flying global civil subsonic air traffic may cause anthropogenic climate changes by an increase of ozone and cloudiness in the upper troposphere, and by an enhanced greenhouse effect. The absolute emissions by air traffic are small (a few percent of the total) compared to surface emissions. However, the greenhouse effect of emitted water and of nitrogen oxides at cruise altitude is potentially large compared to that of the same emissions near the earth's surface because of relatively large residence times at flight altitudes, low background concentrations, low temperature, and large radiative efficiency. Model computations indicate that emission of nitrogen oxides has doubled the background concentration in the upper troposphere between 40°N and 60°N. Models also indicate that this causes an increase of ozone by about 5–20%. Regionally, the observed annual mean change in cloudiness is 0.4%. It is estimated that the resultant greenhouse effect of changes in ozone and thin cirrus cloud cover causes a climatic surface temperature change of 0.01–0.1 K. These temperature changes are small compared to the natural variability. Recent research indicates that the emissions at cruise altitude may increase the amount of stratospheric aerosols and polar stratospheric clouds and thereby have an impact on the atmospheric environment. Air traffic is increasing about 5–6% per year, fuel consumption by about 3%, hence the effects of the related emissions are expected to grow. This paper surveys the state of knowledge and describes several results from recent and ongoing research.

1 Introduction

Until fairly recently, the environmental effects resulting from aircraft exhaust emissions have been a minor topic in the general debate on the environment. However, growing awareness on global climate changes has given rise to discussion, especially in Europe and North America, and aircraft emissions are now perceived as a far more relevant issue than a decade ago. Because of the large residence time, low background concentrations and large radiative sensitivity, the main concern comes from the current air traffic near and above the tropopause, polewards of 40° latitude. The most important emissions are nitrogen oxides, water vapour, and sulphur dioxide. In particular, emissions of nitrogen oxides at high altitudes are considered to possibly have marked effects on the formation or destruction of ozone, depending on deposition altitude. The contributions from water vapour emissions on cloudiness and climate change are also under discussion. More recently, in view of our understanding of the origin of the Antarctic "ozone hole" (WMO, 1992), the potential effect of chemical reactions on the surface of particles (heterogeneous reactions) originating from water, sulphur or soot emissions are gaining increasing attention.

In the early 1970s, concern about the potential effects of emissions from a proposed fleet of supersonic transport (Harrison, 1970; Johnston, 1971; Crutzen, 1972) lead to an extensive study (CIAP, 1975). Partly because of the outcome of this study but mainly because of economical reasons, it was decided at that time that such a fleet of SST (supersonic transport) aircraft should not be built. Recently, there has been renewed interest in the development of faster aircraft for international passenger flight. The question how far such a system is environmentally acceptable triggered the High-Speed Research Program by NASA in 1990 (Watson *et al.*, 1990; Johnston *et al.*, 1991; Douglass *et al.*, 1991; Prather *et al.*, 1992). In Germany, the environmental impact of a hypersonic space transport system SÄNGER was investigated and shown to be small by Brühl *et al.* (1991) and Graßl *et al.* (1991). In 1990, an international colloquium provided an overview on the state of knowledge with respect to the impact of global air traffic on the atmosphere (Schumann, 1990). Since then, several related research programmes have been initiated. This paper surveys the knowledge in this rapidly evolving research area.

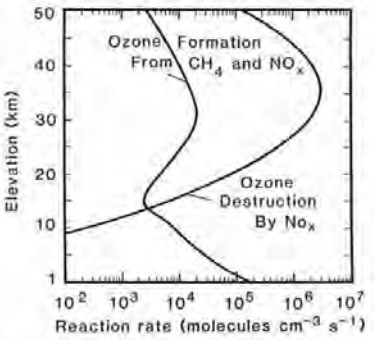

Fig. 1. Ozone formation from the smog reactions based on methane and nitrogen oxides (45° latitude, spring) (Douglass *et al.*, 1991)

Nitrogen oxide NO_x ($NO + NO_2$) plays a major role in the chemistry of tropospheric ozone. In regions of high NO_x concentrations, a photochemical sequence initiated by the reaction of methane (CH_4) and carbon monoxide (CO) with hydroxyl (OH) leads to ozone (O_3) production, while in regions of low NO_x concentrations, such as over ocean areas, the reaction of CH_4 and CO with OH initiates a sequence that destroys O_3 (Isaksen and Hov, 1987; Crutzen, 1988). Hence, additional NO_x enhances ozone in the free troposphere. In the stratosphere, nitrogen oxides as well as hydrogen oxides (HO_x) and in particular chlorine oxides (ClO_x) tend to deplete ozone. As shown by Hidalgo and Crutzen (1977), Johnston *et al.* (1989) and others, using 1D models, NO_x emissions enhance ozone depletion above about 12–14 km but produce ozone below that level, see Fig. 1 (Douglass *et al.*, 1991).

Injection of NO_x by air traffic in the stratosphere may reduce the efficiency of the HO_x and ClO_x catalytic cycles by the homogeneous formation of nitrous acid (HNO_3) and chlorine nitrate ($ClONO_2$). Hence, emissions of nitrogen oxide from air traffic has the potential to reduce ozone destruction caused by increasing levels of chlorofluorocarbons. Johnston *et al.* (1989) found that this effect is small for present levels of ClO. However, the previous models ignored the potential impact of heterogeneous reactions on the surface of stratospheric aerosols and polar stratospheric clouds (PSCs) (Crutzen and Arnold, 1986; Brasseur *et al.*, 1990; Brasseur, 1992; Granier and Brasseur, 1992; McElroy *et al.*, 1992).

The stratospheric aerosol layer contains droplets of aqueous sulphuric acid solution (about 75% H_2SO_4 by weight%). In the absence of sunlight, NO_x is converted into dinitrogen pentoxide N_2O_5. The heterogeneous reaction

$$N_2O_5\,(\text{gas}) + H_2O\,(\text{liquid}) \rightarrow 2\,HNO_3\,(\text{gas}) \qquad (1)$$

on the sulphuric acid aerosol surface converts N_2O_5 and thereby NO_x into the less reactive nitric acid. Hence, this reaction is likely to reduce the ozone depletion effect of NO_x-emissions from high-flying air traffic (Rodriguez *et al.*, 1991; Kinnison and Wuebbles, 1991; Bekki *et al.* 1991; Weisenstein *et al.*, 1991). Dentener and Crutzen (1993) show that such heterogeneous reactions are also important on tropospheric aerosol and water droplets.

PSCs are believed to consist of either HNO_3–H_2O (nitric acid trihydrate, NAT, or metastable nitric acid dihydrate, NAD) or H_2O particles, type-I, and type-II PSCs, respectively (Hanson and Mauersberger, 1988; Arnold, 1992, Hanson, 1992). Type-I PSCs can form at higher temperature than type-II PSCs. The threshold temperature, T(NAT), below which NAT aerosol can exist (in equilibrium) ranges from about 206 K at 10 km altitude to about 191 K at 25 km (Schlager *et al.*, 1990). Because T(NAT) is markedly larger than the threshold temperature of water ice existence (by about 1 K at 10 km altitude and by about 12 K near 25 km altitude), type-I PSCs are more prevalent and thus can have a greater effect on the atmosphere. Heterogeneous reactions on the surface of such PSCs may convert $ClONO_2$ and hydrogen chloride (HCl) to reactive ClO_x which possibly destroys ozone. PSCs also form a sink for NO_x which otherwise moderates the ozone destruction cycles.

The region where NAT particles can be formed increases with decreasing temperature, increasing humidity and increasing concertration of HNO_3 (Peter *et al.*, 1991; Hofmann and Oltmans, 1992; Arnold *et al.*, 1992). Increases in CO_2 concentrations may cause cooling of the lower stratosphere and more PSCs and hence more ozone depletion in the Antarctic and eventually also in the Arctic (Austin *et al.*, 1992). Hence, aircraft emissions may also enforce ozone destruction in the coldest regions of the stratosphere. The overall consequences of such heterogeneous reactions have not yet been investigated in detail with respect to emissions from present subsonic air traffic.

Over the industrialized continents, a strong increase of tropospheric ozone has been observed. Wege (1992) and Vandersee *et al.* (1993) observed an increase of upper tropospheric ozone of about 2%/year, at least until the mid-1980s see, e.g., Fig. 2. Volz and Kley (1988) deduced

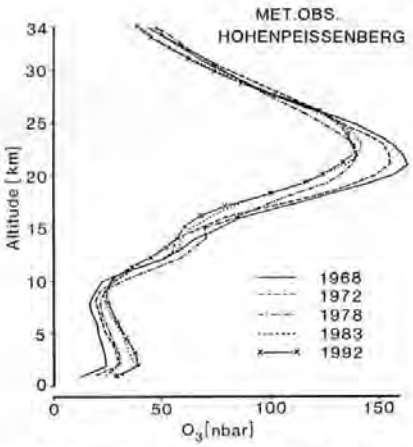

Fig. 2. Annual mean vertical profiles of ozone partial pressure in nbar versus altitude in km for the years 1968 to 1992 at Hohenpeißenberg, Southern Germany (provided by Meteorol. Observatory Hohenpeißenberg, German Weather Service)

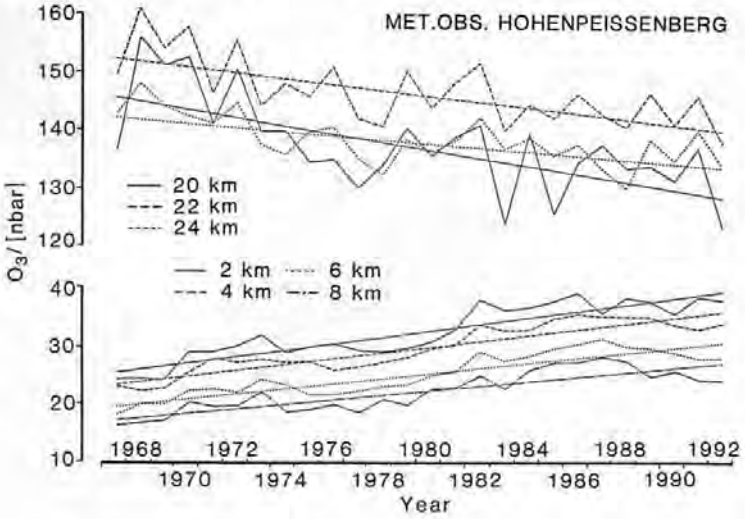

Fig. 3. Annual mean ozone pressure in nbar versus time at various altitudes in km at Hohenpeißenberg, Southern Germany (provided by Meteorol. Observatory Hohenpeißenberg)

more than a doubling compared to the pre-industrial era. Increases in upper tropospheric ozone of about 1.5% per year have also been observed at other ozone stations (WMO, 1992). This trend is considered to be consistent with increases of nitrogen oxide (Logan, 1985; Brühl and Crutzen, 1988). Recent measurements, see Fig. 3 (Vandersee, 1993), suggest an interesting saturation or even decline in the tropospheric ozone trend in the last 5 years at Hohenpeissenberg. The significance of this change in trend and its reasons are not yet clear (see Section 7). Figure 2 shows a reduction of ozone of about 0.5%/year (Wege, 1992) in the stratosphere. Satellite measurements of total column amount of ozone also show a decrease of stratospheric ozone by about 0.5–0.8%/year near 50°N (Stolarski *et al.*, 1991).

The changes in the ozone concentration have environmental effects because ozone is a greenhouse gas (Lacis *et al.*, 1990). On the other hand, ozone is important for the production of OH which is responsible for the removal of greenhouse gases (e.g., methane) and other pollutants (Crutzen and Zimmermann, 1991). Moreover, the ozone layer protects the biosphere from the UV radiation. Increased tropospheric ozone may cause damage to both animal and plant life (UNEP, 1991).

The other potentially important emission is that of water vapour, mainly because of its direct or indirect (by cloud-forming processes) impact on the radiation budget of the atmosphere. It is known that thin cirrus clouds of large ice particles in the upper troposphere at low and mid-latitudes enhance the "greenhouse effect". An increase in cirrus cloud coverage by a few percent might have the same effect as a doubling of the amount of CO_2 (Liou, 1986; Liou *et al.*, 1990). Contrails from aircraft enhance the formation of high-level clouds (Schumann and Wendling, 1990).

The purpose of this paper is to review the present knowledge of the impact of high-flying civil subsonic air traffic on the atmosphere in terms of environmental and climatic aspects. The paper excludes effects of air traffic in the vicinity of airports.

2 Air traffic and emissions

Global air traffic of scheduled services has grown from 940×10^9 passenger-km in 1978 to almost 1700×10^9 passenger-km in 1988, 45% of the total being international traffic (Nüßer and Schmitt, 1990). The non-scheduled traffic adds about 170×10^9 passenger-km for the year 1988. The annual growth rate in the decade between 1978 and 1988 was 6.1%. ICAO expects an increase of worldwide scheduled passenger traffic at an annual growth rate of about 4% for domestic traffic and 6% for international traffic in the years until 2001, with the strongest increase (up to 8.5%/year) for international traffic in the Asia/Pacific region.

Data by the International Energy Agency of 1991, see Fig. 4, show that the world demand of aviation fuel increased from 117 Mt (million tons) in the year 1977 to

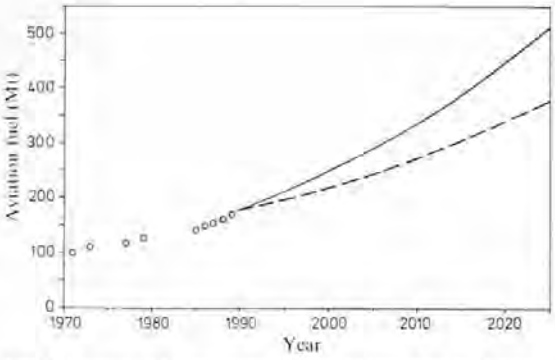

Fig. 4. Aviation fuel versus time. Data up to 1989 from the International Energy Agency (1991). Extrapolations according to Kavanaugh (1988) with 2.2% per year in a low fuel scenario and with 3.6% up to 2000 and 2.9% thereafter in a high fuel scenario

167 Mt in the year 1989, i.e., by about 3% per year. Hence, the increase rate in terms of fuel consumption is roughly half the rate of passenger transport performance. Kavanaugh (1988) estimates that commercial jets burn about 70% of all jet fuel, military jets consume 24% and business and turbo-props the remaining 6%. These numbers are roughly consistent with information from G. J. Bishop, Shell International Petroleum Co., London (personal communication, 1991, 1993), reporting jet fuel consumption by civil airlines in the western world in 1990 of approximately 125 Mt and estimating an additional military share of 20% of total volume. For 1992 he found that the military share decreased to 7% of total world jet fuel demand. For the future, Kavanaugh (1988) estimates the jet fuel use to grow by 2.2% annually from the year 1990 to 2025 in a "low fuel estimate" scenario, but to grow 3.6% per year from 1990 to 2000 and then moderate to 2.9% in a "high fuel estimate", depending on various assumptions. These estimates imply increases by a factor of 1.4–1.6 in 2005 and by 2.1–2.9 in the year 2025 relative to 1990. Greene (1992) expects a considerably lower growth rate of between 0.8 and 1.8% for the American commercial aircraft fleet in 1989–2010.

Hence, although air traffic is expected to increase strongly, the fuel consumption is unlikely to double before 20 years from now. However, it appears reasonable that most of the increase will be related to high-flying long-distance traffic. The lower increase of fuel consumption when compared to the increase in the number of passenger-kms is related to increases in the fuel efficiency of aircraft. Within the last 30 years, the specific fuel consumption per passenger-km decreased by about 45% (Grieb and Simon, 1990).

When the fuel is burnt with air in the jet engines, the exhaust gases cause emissions into the atmosphere. For complete combustion, the emissions would contain mainly carbon dioxide (CO_2) and water vapour (H_2O). The emission index (emitted mass of pollutant per unit mass of fuel) for these emissions depends solely on the stoichiochemical composition of the kerosene fuel. The carbon content is about 86% by mass. The mean molecular weight is 164 (Prather *et al.*, 1992, p. 98). Other emissions of much smaller amount are nitrogen oxides (NO_x) (Miller and Bowman, 1989) of varying composition (NO_2 fraction decreasing from 18% at idle to less than 5% at more than 30% engine power setting, according to Spicer *et al.*, 1990), carbon monoxide (CO), methane (CH_4), non-methane hydrocarbons (HC), and soot particles (mainly carbon) and other particles acting as condensation nuclei (CN) for cloud formation. One distinguishes between CN and CCN (cloud condensation nuclei). CCN form cloud nuclei at small supersaturation (less than 1%), whereas CN require large supersaturation. Pitchford *et al.* (1991) measured large concentrations of CN of up to 300 000 cm^{-3} in the exhaust plume of a Sabreliner. They found that background particles are much more active as CCN than exhaust particles of the same size (ratio of CCN to CN less than 1%), but Whitefield *et al.* (1993) found that 30–40% of the combustion aerosols generated in laboratory burners can be considered as CCN. Most of the aerosols in jet exhausts are found to have diameters between 0.01 and 0.1 μm (Pitchford *et al.*, 1991; Hagen *et al.*, 1992). Black carbon soot aerosol is also of radiative importance, because it has a high absorption cross section (Pueschel *et al.*, 1992).

As a minor emission, the exhaust contains sulphur dioxide (SO_2), because aviation fuel is allowed to contain up to 0.3% by weight (wt%) of sulphur (IATA guidance material for aviation turbine fuels, Amendment No. 1, 14 Nov. 1988, International Air Transport Association, Montreal, Canada). According to Shell, London (personal communication, 1991), fuels can vary from very low sulphur contents (< 0.01 wt%), if they have been manufactured by a hydroprocessing route, to levels around 0.2 wt%, if they have been chemically processed. From 1978 to 1991, the average sulphur content in fuel samples was between 0.050 and 0.055 wt% (Dickson, 1989, 1992), with very few samples exceeding 0.1 wt%. In an analysis of 53 Jet-A-fuel samples the average sulphur content was 0.042 wt% (Prather *et al.*, 1992). As confirmed by Shell, the average sulphur content of jet fuel worldwide is around 0.04–0.05 wt%; resulting in 0.8–1 g SO_2 per kg burnt fuel.

Very little information exists on the composition of the HC. The standard method of exhaust analysis for organic compounds is only a measure of "total unburnt hydrocarbons" as measured by a flame ionization detector. The specific compounds are of interest because the exhaust organics vary substantially in their chemistry. Such organic compounds may be important in converting NO_x into reservoir gases and in contribution to ozone formation, in particular in the stratosphere. Any technical development leading to reduced NO_x emissions might lead to increasing amounts of unburnt hydrocarbons. Spicer *et al.* (1990) reviewed earlier investigations and reported on measurements in the exhaust of two military engines, the F101, used on B-1B aircraft, and the F110, used for F-16C and F-16D aircraft. They found that many of the hydrocarbons consist of methane, part of which enters the engine with the combusted air, in particular for low power settings. The content of non-methane hydrocarbons, mainly alkanes, decreases with the load factor of the engine. The organic compounds in the two engines reached up to 1.44 g/kg fuel in the idle state and decreased below 0.2 g/kg fuel at power settings above 63%.

For NO_x, published emission indices vary considerably. This is mainly due to the fact that measured emission indices are obtained for surface pressure conditions while the emissions at cruise height are much smaller. Measurements in altitude test chambers are being made to determine these differences. The emission index for NO_x is usually specified as if all NO_x were in the form of NO_2. Given values of the emission index in g(NO_2)/kg(fuel) are 10.9 (Kavanaugh, 1988), 9.3 around 8 km and 14.4 for long-range cruise at an altitude between 10 and 11 km (Ko *et al.*, 1992; the differences reflect different mixes of aircraft), 11.6 (Johnson *et al.*, 1992, McInnes and Walker, 1992), 15 (Reichow, 1990), and 18 (Beck *et al.*, 1992) based on the data compiled by Lecht *et al.* (1986). Even larger variations are found with respect to emission indices for CO: 14.6 (Johnson and Henshaw, 1991), 4 (Reichow, 1990), and 0.7–2.5 (Nüßer and Schmitt, 1990), based on

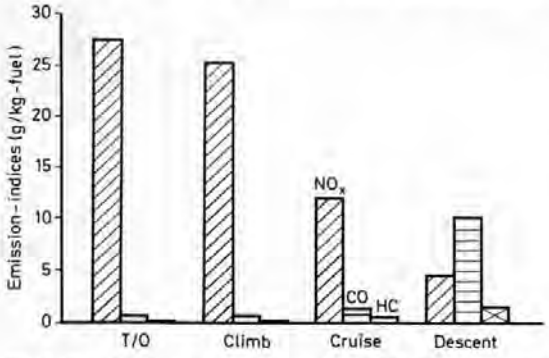

Fig. 5. Calculated emissions indices of a B747-400 with a CF6 engine at take/off (T/O), during climb, cruise and descent, for NO_x, CO, and HC (from Deidewig, 1992)

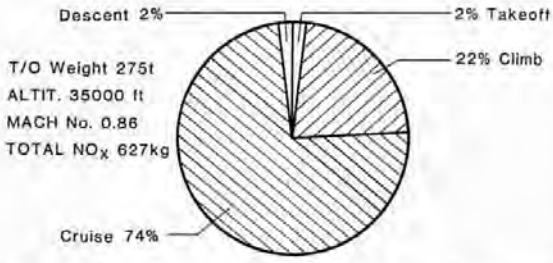

Fig. 6. Percentage NO_x emissions of a B747-400 with a CF6 engine during take/off (T/O), climb cruise and descent (From Deidewig, 1992)

Lufthansa estimates. For HC, the variations are 0.05–0.7 (Nüßer and Schmitt, 1990), 1.5 (Reichow, 1990), and 2.6 (Johnson and Henshaw, 1991). With respect to soot the figures are 0.007–0.03 (Nüßer and Schmitt, 1991), 0.014 (Reichow, 1990), and 0.001–0.03 (Pitchford *et al.*, 1991).

Deidewig (1992) evaluated the emissions for the aircraft types B737-300, B737-500, A310-300, and B747-400, propelled by the engines CFM56-3 and CF6-80C2. For various typical missions, he computed the emissions of NO_x, CO, and HC in the atmosphere along the flight route. For the B747-400, his results are repeated in Figs. 5 and 6. We see that the emission indices of NO_x vary between 8 and 12 at cruise, where between 31% and 74% of the total NO_x emissions take place. Larger specific emissions occur in other parts of the routes, which are, in particular, important for short range missions. Note that this analysis applies to a specific set of engines with NO_x emission indices that are lower than average.

Hoinka *et al.* (1993) recently analysed the traffic over the North Atlantic in the region shown in Fig. 7, i.e., between 10°W and 60°W and 45°N and 65°N. Based on data of the Civil Aviation Authority (CAA), they find that on average 512 aircraft per day (sum of both directions) passed that region during the period from 1989 to 1991.

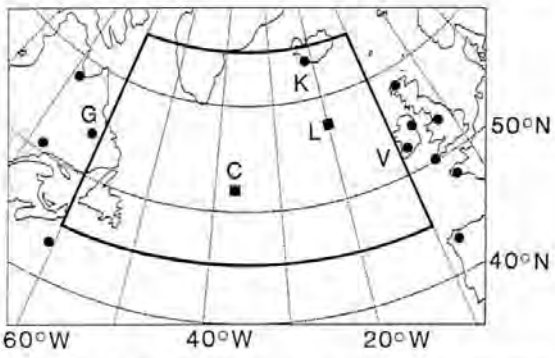

Fig. 7. Map of the Northern Atlantic showing the area for which Hoinka *et al.* (1993) performed an analysis of traffic and tropopause statistics. The *dots* (*squares*) indicate land (ship) stations where radiosondes are launched twice a day

Based on the fleet composition as given by the CAA and estimates for the individual engines used (Lecht *et al.*, 1986), they determine the total emissions and fuel consumptions from which one obtains the following average emission indices for the whole fleet: 17.5 for NO_x, 0.6 for HC, and 1.5 for CO. The amount of fuel used by the average aircraft in that region, with an average flight path length of 2730 km, amounts to 32.4 t.

Based on the various sources and arguments, Table 1 contains what we believe are best estimates of emission indices. They apply to long-distance cruise conditions. The table also contains an estimate of current (as of 1990) emissions from air traffic. In this table, the most important uncertainty is that of NO_x emissions (which are qualified as if all emissions were NO_2), for which we have selected the upper bound of the various estimates.

With respect to the spatial distribution of the emissions, previous estimates have been compiled in Schumann and Reinhardt (1991), Ko *et al.* (1992) and Beck *et al.* (1992). These data are not repeated here, because several studies are in progress with the objective to produce a refined data base (Prather *et al.*, 1992; Lister and Gardner, 1993).

As a preliminary result from these studies, a 3D data base of emissions has been provided by McInnes and Walker (1992). These data still contain considerable uncertainty, because the data are based on traffic statistics for scheduled flights and cargo only. Hence, the resultant fuel consumption accounts for only about 50% of the total fuel consumption. The results have been scaled by the factor 2 to obtain an estimate of the total emissions. For illustration purposes, Fig. 8 shows the distribution of the source of NO_x emissions per unit volume and unit time at an altitude of about 12 km (190 hPa). We see clearly that the source at this altitude is large, in particular, along the main flight corridors between the northern continents with peaks over North America, Europe and the North Atlantic.

Table 1. Estimated emission rates and comparison. Best estimate emission indices are listed together with the present range of uncertainty in brackets. The corresponding emissions are given in units of Mt/year (10^{12} g year^{-1}). References for comparable emissions: (1) International Energy Agency (extrapolated from 3095 Mt/year for 1989). (2) German Bundestag (1991), p. 151 ($5.7 \cdot 10^9$ t C/year in 1987 from anthropogenic sources), (d) German Bundestag (1991), p. 156, (4) p. 147, (5) p. 170, (6) p. 168, (7) p. 168. (8) Hough (1991). (9) Hofmann (1991). (10) Hameed and Dignon (1992).

Emission	Emission index (g pollutant per kg fuel)	Emission rate 1990 in Mt/year	Comparable emissions in Mt/year	Comparable emissions source	Reference
Fuel	1000	176	3140	Total consumption of petrol	1
CO_2	3150	554	20900	Burning of fossil fuels	2
H_2O	1260	222	45	Methane oxidation in the stratosphere	3
			525000	Evaporation from earth surface	4
NO_x (as NO_2)	18 (7–20)	3.2	2.9 ± 1.4	Flux from stratosphere	5
			90 ± 35	All anthropogenic sources	5
CO	1.5 (1.5–10)	0.26	600 ± 300	CH_4 oxidation	6
			1490	All anthropogenic sources	7
HC	0.6 (0.2–3)	0.1	90	Anthropogenic emissions at the earth's surface	8
Soot (C)	0.015 ± 0.014	0.0025			
SO_2	1 (0.02–6)	0.176	0.0625	Rate required to sustain background aerosol in the lower stratosphere	9
			134	Total from fossil fuel combustion	10

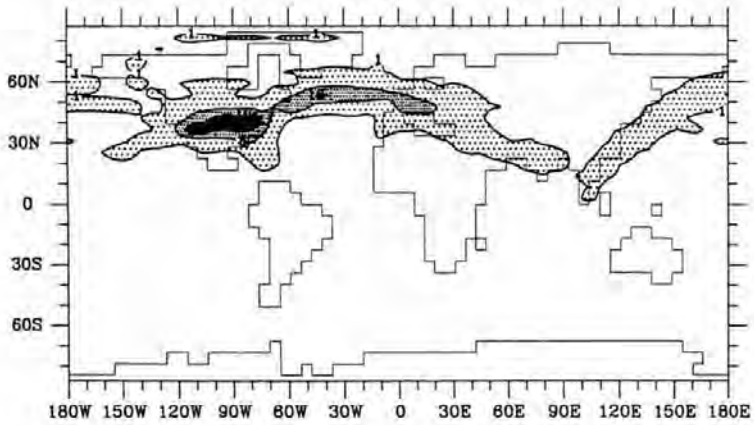

Fig. 8. Source of NO_x due to air traffic emissions at an altitude of about 12 km (at pressure level 190 hPa) in the T21 grid version of the ECHAM code. The contours correspond to emissions of 1, 5 and 10 in units of 10^{-10} kg(NO_2)/(kg(air)s). The emissions exceed 10 such units within the *black areas*

3 Some relevant properties of the atmosphere

The concentrations which result from the emissions are larger the larger the product of emission mass rate and residence time and the smaller the mass of atmosphere over which the emissions are dispersed. (Mass specific concentrations are converted to volume specific concentrations by multiplication with the molecular weights $M_i = 44, 18, 46, 18, 64$, and 29, of the individual gases CO_2, H_2O, NO_2, CO, SO_2, and air, respectively). Hence, the concentration change can be estimated from

$$\text{Volumetric concentration change} = \frac{\text{emission rate} \times \text{residence time} \times M_{air}}{\text{mass of atmosphere} \times M_i}. \quad (2)$$

The residence time and the relevant mass of atmosphere depend strongly on the emission altitude and on the manner by which the emitted quantities are mixed and removed from the atmosphere.

As described in Brasseur and Solomon (1986), Fabian (1992) and others, the atmosphere is structured into various layers. The lower troposphere contains roughly 80% of the mass of the atmosphere. In the troposphere, the temperature decreases with height (the potential temperature increases slightly), so that this layer is only weakly stably stratified. In this range, vertical mixing is quite strong and cloud systems contribute to deposition of soluble quantities. Above the troposphere, i.e. above the tropopause, within the stratosphere, the temperature increases with height. Therefore, this layer is stably stratified with much weaker vertical mixing. As a consequence of these properties and of the conversion into soluble forms, nitrogen oxides have a residence time of 1–4 days in the troposphere. For inert tracers emitted into the stratosphere, the residence time quickly increases with height above the tropopause and reaches values in excess of 1 year (Fabian, 1992). For current air traffic at altitudes below 13 km, the residence time is probably below 6 months (see Fig. 9 of Brühl *et al.*, 1991). The altitude of

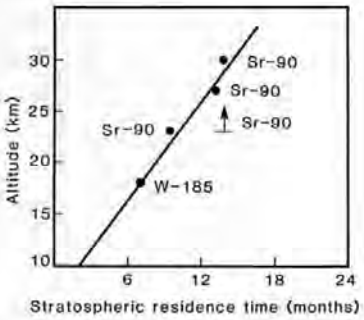

Fig. 9. Height-dependent residence time in the stratosphere in units of months, deduced from radioactive tracers (from Brühl *et al.*, 1991)

the tropopause is highly variable from day to day and from season to season (Hoerling *et al.*, 1993, Hoinka *et al.*, 1993). It amounts on average to about 8 km over the polar regions and to about 16 km at the equator. Hence, the residence times of aircraft emissions depend strongly on whether the aircraft flies above or below the tropopause.

The residence time also strongly depends on the latitude, because the lower stratosphere exhibits mean rising motion near the equator and, to a first approximation, mean sinking motion elsewhere (Plumb and Mahlman, 1987). The residence time also varies strongly at midlatitudes because of strong tropospheric–stratospheric exchange (Vaughan, 1988; Ebel *et al.*, 1991; Rood *et al.*, 1992; Hoerling *et al.*, 1993). The polar jet stream is important because of sinking motion (50–500 m per day) near or poleward of its core and rising motion of its equatorward side (Mahlman, 1973; Schoeberl *et al.*, 1992). Hoerling *et al.* (1993) diagnose the cross-tropopause mass flux globally for January 1979. They find that the mass flux from the troposphere to the lower stratosphere between 50°N–70°N is as intense as that over the tropics, while the same flux is rather small over the poles. This suggests that material deposited in the stratosphere or near the tropopause poleward of the jet core would have shorter residence time than material injected equatorward of the jet core. Jackman *et al.* (1991) show that ozone depletion from NO_x injections of stratospheric supersonic aircraft, as computed in 2D models, depends strongly on the stratospheric/tropospheric exchange rate by the mean circulation. The computed ozone loss is reduced when the exchange rate is increased.

Reichow (1990) estimated that Lufthansa consumes about 17–20% of the fuel above the tropopause. Havlik (1988) analyzed radiosonde data for the year 1970 for 00 and 12 UTC. He determined the height of the tropopause (i.e. the lowest altitude above which the vertical temperature gradient was about -2 K/km over an interval of at least 2 km). From his data, aircraft at 250 hPa or about 33 000 ft (10.8 km) altitude most of the time fly in the stratosphere when cruising north of 50°N. Ko *et al.* (1992) use zonally averaged climatologies and estimated the fuel consumption in the stratosphere for the 1987 subsonic fleet assuming an average flight altitude of 37 000 ft. They conclude that 48% of the 1987 northern hemisphere fuel burn occurs in the stratosphere and 52% in the troposphere, with strong seasonal variations. These numbers are likely to change when more accurate emission inventories become available (see also Hoinka *et al.*, 1993, and Section 5).

4 Global concentration changes due to current aircraft emissions

Table 1 lists the emitted masses of various pollutants and gives other emission rates for comparisons. We see that nearly 6% of all petrol is used for aviation. This is quite a large proportion. However, because of many other sources, it contributes only 2.6% of all CO_2 from the burning of fossil fuels. The water vapour emission rate of aircraft is very small (0.04%) compared to the amount of water evaporating from the earth's surface. However, it is large when compared to the amount of water vapour released into the stratosphere by methane oxidation.

Table 2 gives an overview of various sources of NO_x. Aircraft cause the only direct anthropogenic emissions in the upper troposphere. Other sources are those from lightning and stratospheric downward flux. Note that there is much disagreement regarding the source of NO_x from lightning (Jacob, 1991). Liaw *et al.* (1990) estimates that the production rate of nitrogen oxides amount to 152 ± 60 Tg N year^{-1}, part of it being washed out with the rain. Lightning sources represent an important uncertainty for the assessment of aviation impact (Beck *et al.*, 1992). The upward flux from surface emissions will be large only over the continents, in cyclones, and in the tropics where strong convective events quickly transport the pollutants from the surface up into the upper troposphere (Ehhalt and Drummond, 1988). The aviation emissions are large compared to the flux of NO_x from the stratosphere into the troposphere.

The aviation contribution to all anthropogenic NO_x sources is about 3%. The emissions from air traffic grow faster than the other anthropogenic emissions which will increase the relative importance of the former. For

Table 2. Global sources of nitrogen oxide and their strengths, in units of Mt (N)/year (10^{12} g N year^{-1}). 14 kg N $\cong$ 46 kg NO_2. For 1986, Hameed and Dignon (1992) estimate the total emissions from fossil fuel combustion to be 24 Mt(N)/year

Emission	Ehhalt and Drummond (1988)	Penner *et al.* (1991)	Hough (1991)	German Bundestag (1991), p. 170
Fossil fuel burning	13.1–28.9	22.4	21	20 ± 7
Biomass burning	5.6–16.7	5.8	8	7 ± 3
Soil microbial activity	1–10	10	5	12 ± 6
NH_3 oxidation	1.2–4.9			3 ± 2
Lightning discharges	2–8	3	8	5 ± 3
Flux from stratosphere	0.3–0.9	1.0		1.0 ± 0.5

example, the global emissions of nitrogen from fossil-fuel combustion increased from 18 Mt N/year in 1970 to 24 Mt/year in 1986, i.e. at a rate of 1.8%/year (Hameed and Dignon, 1992). Obviously, various reduction measures with respect to emissions at the earth's surface (e.g. from car traffic) become effective (Amann, 1990).

The amount of emitted HC appears to be small compared to anthropogenic sources. Also the CO contributed from aviation is very small compared to the budget of CO. It reaches only 0.03% of all anthropogenic sources. The CO concentration may be reduced in spite of the aircraft emissions; this is caused by a greater oxidation of CO due to enhanced concentrations of OH as shown by Beck *et al.* (1992).

The absolute amount of soot induced by aircraft into the atmosphere is certainly much smaller than those from other sources. Moreover, this class of emissions has already been reduced considerably in the past. However, soot may still contribute significantly to the CN concentration in the lower stratosphere (Pitchford *et al.*, 1991). Pueschel *et al.* (1992) measured the background soot concentration above and below the tropopause. In the stratosphere, they find an average concentration of 0.6 ng/(standard m)^3. In the troposphere, the soot concentration is larger by a factor of 10–50. From estimated emissions and a lifetime of 1 year, they conclude that current aircraft emissions may be the predominant source of the present stratospheric soot concentrations. They also find that planned supersonic transport systems may double the stratospheric soot concentration, reduce the aerosol single scattering albedo by 1%, which is of climatic relevance, and double the soot surface that is available for heterogeneous chemistry.

The emission of sulphur from aviation is much smaller than from surface emissions and negligible in terms of the resultant acid rain etc., but may be important if emitted at high altitudes. Hofmann (1991) reported observations which indicate an increase of non-volcanic stratospheric sulphuric acid aerosol of about 5% per year. He suggests that if about 17% of the Northern Hemisphere air traffic takes place directly in the stratosphere and if a small fraction of other emissions above 9 km enters the stratosphere through dynamical processes then the jet fleet appears to represent a large enough source to explain the observed increase. He calls for the use of aviation fuel which is essentially sulphur-free. On the other hand, Bekki and Pyle (1992) conclude from a model study that, although aircraft may represent a substantial source of sulphate below 20 km, the rise in air traffic is insufficient to account for the observed 60% increase in large stratospheric aerosol particles over the 1979–1990 period. Particles generated from SO_2 may also contribute to nucleation particles (Arnold *et al.*, 1992). Whitefield *et al.* (1993) find a positive correlation between sulphur content and CCN efficiency of particles formed in jet engine combustion. Bekki and Pyle (1993) discuss the impact of SO_x emitted by a fleet of high-speed civil transport on the aerosol surface area, the number of optically active particles and heterogeneous chemistry.

Tables 3 and 4 show equilibrium concentrations which are computed using Eq. (2). Here, we follow Fabian (1990) who presented similar estimates. Note that these computations neglect any chemical transformations. In Table 3, we present an update of such computations for the stratosphere assuming that 20% of the fuel (total 176 Mt/year) is burned within 10% (i.e. only the northern stratosphere) of the atmosphere's mass of 53×10^8 Mt total (Fabian, 1990). The residence times and background concentrations are as estimated in Schumann and Reinhardt (1991). For the troposphere (Table 4), it is assumed that 80% of all fuel is burnt and distributed over 80% of the atmosphere's mass (i.e. below 200 hpa).

Table 3. Concentration increases due to aviation emissions mixed over the northern stratosphere

Emission product	Residence time (years)	Emission rate in Mt/year	Mean concentration increase	Background concentration at 14 km
CO_2	10	111	1.4 ppmv	354 ppmv
H_2O	0.5	44	0.07 ppmv	5–20 ppmv
NO_x	0.5	0.63	0.4 ppbv	0.2–0.5 ppbv
SO_2	0.5	0.035	16 pptv	30–100 pptv

Table 4. Concentration increases due to aviation emissions mixed over the whole troposphere

Emission product	Residence time	Emission rate in Mt/year	Mean concentration increase	Background concentration at 8 km
CO_2	10 years	443	0.7 ppmv	354 ppmv
H_2O	9 days	177	0.002 ppmv	20–400 ppmv
NO_x	4 days	2.5	4.1 pptv	10–100 pptv
SO_2	5 days	0.14	0.21 pptv	30–150 pptv

The contributions of emissions from air traffic are obviously significant with respect to NO_x. The changes may reach 10–100% of the background concentrations both in the upper troposphere and in the lower stratosphere, even when the emissions are assumed to be mixed over such large volumes. Also, the contribution of air traffic to SO_2 is significant in this global mixing analysis, particularly in the lower stratosphere. On the other hand, such calculations show a rather small effect of emitted water vapour. Larger effects are to be expected regionally.

5 Traffic, dispersion and concentration changes over the Northern Atlantic

Duclos *et al.* (1991) reported a North Atlantic traffic analysis using traffic data as provided by the CAA and presented forecasts for the year up to 2010. Figure 10 shows the annual number of aircraft movements over the North Atlantic in the past and as estimated for the future. We see that these numbers increased from 120 000 in the year 1976 up to 206 100 in 1990 and the study predicts about 356 000 movements in the year 2005. This corresponds to about 560 aircraft movements per day in 1990, increasing to nearly 1000 by 2005.

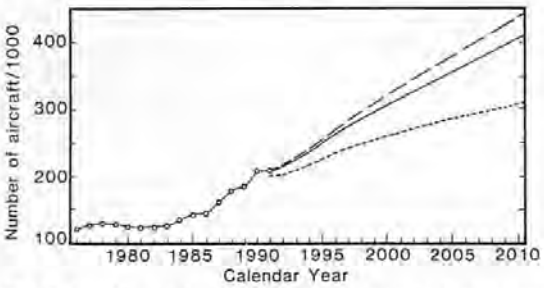

Fig. 10. North Atlantic traffic forecasts showing aircraft movements 1976 to 2010. The extrapolations apply for various traffic scenarios. *Full curve with circles*: past traffic, *full curve*: best estimate, *long and short dashed*: high and low limit

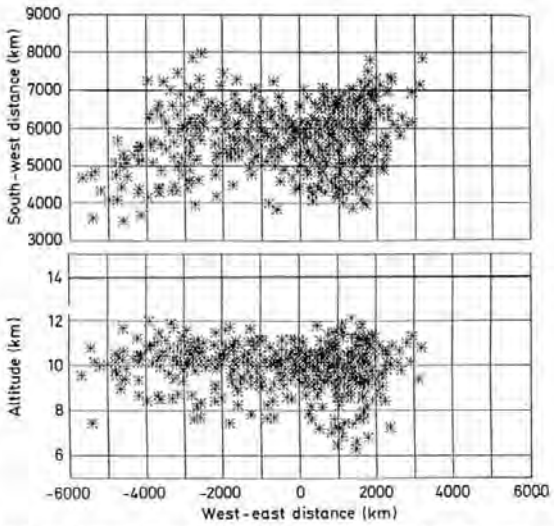

Fig. 12. Particle positions as released along the 40 aircraft tracks in June 1991 after 24 h dispersion along the trajectories, computed using the "Europa model" of the DWD. The plots show the distribution in a horizontal plane (top) and a vertical west-east plane (bottom)

As mentioned before, Hoinka *et al.* (1993) also analysed the traffic in the region depicted in Fig. 7. In addition, they determined the height of the tropopause from weather analysis data as provided by the European Centre for Medium Range Weather Forecasts (ECMWF) in Reading, UK. By correlating both data sets, they found that about 44% of all fuel is burnt above the tropopause in the lower stratosphere and 56% in the troposphere in that region. In February, the stratospheric share reaches about 75%.

Schumann and Reinhardt (1991) reported on trajectory analysis of emissions which are emitted along the path of an airliner, according to Lufthansa flight dispatch data and meteorological data from the German Weather Service (see Fig. 11 for example). A statistical analysis of the results (see Fig. 12) shows that the standard deviation σ in the vertical amounts to 0.729 km at the time of tracer release (due to variation in flight altitudes), 0.860 km after 12 h, 1.014 km after 1 day and 1.194 km after 2 days. The north-south standard deviations are 496, 638, 854, 1303 km after 0, 12, 24 and 48 h, respectively. This corresponds to mean diffusion coefficients $K = 0.5\partial K\sigma^2/\partial t$ of 2.8 $m^2 s^{-1}$ in the vertical, and 4.6 $km^2 s^{-1}$ in north-south direction. The related vertical dispersion extends from about 9 to 11 km within the first day, i.e. over about 2 km. Hence, the emissions from this and the other flights on the same day, which take various routes staggered over about 500–800 km (at 20° W) in north-south direction, become distributed over the flight corridor with a typical cross section of about 1000–1500 km in latitude and about 2 km in height along a distance of about 5000 km. The tracer results also indicate that residence times in excess of several days are unlikely for emissions near the tropopause.

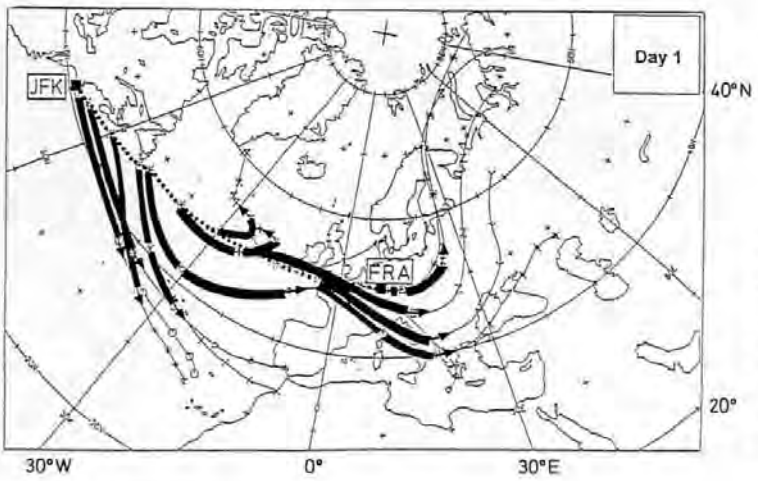

Fig. 11. Forward trajectories of emission products along a North Atlantic eastbound jet aircraft track from New York (JFK) to Frankfurt (FRA) on June 20, 1991 at flight level 10 km. *Dotted line*: flight track, *thick arrowed curves*: horizontal trajectory for 24 h, for day 1 after release. The thin trajectory curves extend up to 3 days. The symbols at the trajectories mark time intervals of 12 h. Computed with the "Europa model" of the DWD (German Weather Service)

Table 5. Concentration surplus in the North Atlantic flight corridor due to air traffic exhaust emissions from 500 aircraft consuming 60 t of fuel per 5000 km mixed over a corridor cross section of 1000 km width and 2 km height. Assumed residence time = 1 day within corridor for all emitted gases

Emission product	Mean concentration increase	Background concentration at 8 km
CO_2	0.02 ppmv	354 ppmv
H_2O	0.02 ppmv	20–400 ppmv
NO_x	97 pptv	10–100 pptv
SO_2	4 pptv	30–150 pptv

Based on these analyses, the mean concentration increase is estimated (see Table 5) assuming an air volume of $1000 \times 5000 \times 2$ km with an air density of 0.35 kg m^{-3}, and a residence time of 1 day for all emissions which originate 500 aircraft consuming 60 t of fuel each in this corridor. The latter number corresponds to the above mean fuel consumption per distance as estimated by Hoinka *et al.* (1993).

One finds that the concentration change of NO_x in the flight corridor can be larger than the background concentration value. The mean concentration increase in SO_2 is still appreciable. On the other hand, the effect of emitted water vapour is small. The emitted water may become important in flight corridors, in regions with long residence times and where the background concentration is small.

Figure 13 shows the increase in NO_x as computed with the global circulation climate model ECHAM (Roeckner *et al.*, 1992; Cubasch *et al.*, 1992), using the NO_x source distribution as illustrated in Fig. 8 and a sink corresponding to a residence time of 10 days (Sausen and Köhler, 1993). The picture shows a zonal cross section along 50°W, i.e. through the North Atlantic flight corridor. The maximum concentration computed in this plane amounts to 125 pptm = 82 pptv. In view of the various assumptions, this value is surprisingly close to the simple corridor estimate given in Table 5. The emissions from the flight corridors over the North Atlantic are mixed rather quickly downwards, adding to material transported from the North American continent. There is relatively little transport upwards but some transport northwards into the lower stratosphere over the Arctic. In the troposphere one finds some mixing in north-south direction. The direct contribution of air traffic emissions to nitrogen oxides in the South Atlantic appears to be very small. More refined simulations are in progress (Schumann *et al.*, 1993).

The simple corridor estimate is also approximately consistent with the analysis of Ehhalt *et al.* (1992) who used a 2D model to study the zonal distribution of NO_x. The model includes vertical transport in the form of eddy diffusivity and deep convection, zonal transport by a uniform mean wind and a simplified chemistry of NO, NO_2, and HNO_3. The model distinguishes between the contributions from various emission sources and is applied to the latitude band 40°–50°N during July. The model predictions of NO compare quite well with experimental observations. Ehhalt *et al.* (1992) conclude that aircraft emissions contribute on average 30% to NO_x concentration in the upper troposphere. The contribution is high, in particular over the North Atlantic (see Fig. 14). Table 5 suggests an even larger percentage contribution which might be caused by differences in the assumed rate of emission. Kasibhatla (1993) used a global chemical transport model and found that fast upward transport of surface emissions are not necessary to explain measured NO in the upper troposphere at northern mid-latitudes.

6 Concentration change in the wake of an airliner

The processes in the aircraft wake control the formation of contrails, the formation of sulphuric particles and CN, and determine the composition of emissions as becoming effective for the global scale. The details of these processes are important for nonlinear conversion processes.

The aircraft wake can be conveniently subdivided into three regimes (CIAP, 1975): the jet, the vortex, and the dispersion regimes. These regimes correspond to the flow dynamics that control the structure and growth of the wakes in the subsequent time interval. In the jet regime,

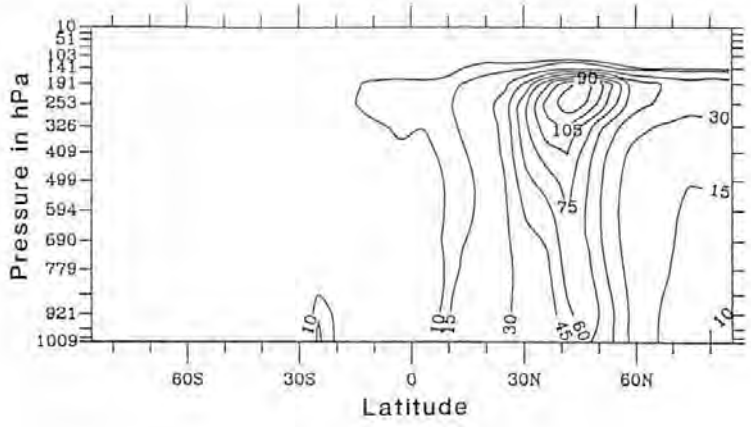

Fig. 13. Increase of NO_x due to air traffic emissions as computed by the ECHAM-T21-model in a zonal plane at 50°W for July as a function of latitude and height (in pressure units). The contours are plotted for 10, 15, 30, 45, . . . , 120 ppt

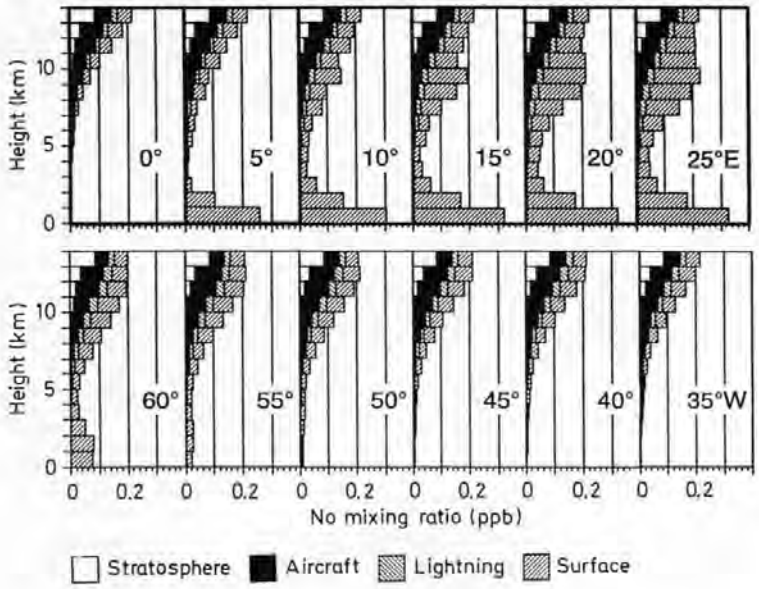

Fig. 14. Vertical mixing ratio profiles of NO over the western North Atlantic (bottom) and over continental Europe (top). The different shadings indicate the contributions from the individual sources. The *black part* defines the contribution from aircraft emissions. From Ehhalt *et al.* (1992)

the engine effluents are initially confined to individual exhaust jets. At the end of the jet regime, the jets merge and are entrained in the roll-up vortex. The following vortex regime persists in a calm atmosphere until the vortices become unstable (Crow, 1970) and break up into a less ordered configuration. Thereafter, the dispersion regime follows, in which further mixing depends on atmospheric shear motions and turbulence. Also, buoyancy from the jet exhaust heat or from differential cooling of contrails by radiation may contribute to this mixing.

Using simple analytical models (Donaldson and Bilanin, 1975; Miake-Lye, *et al.*, 1993) for a B747, one can estimate that the effective cross section of mixing grows in the jet regime within 10 s from about 3 m^2 near the four engine exits to about 3600–5400 m^2 at the end of the jet regime. In the following vortex regime, which lasts for about 2–3 min, the cross section of the trailing vortex pair grows to about 21 000–52 000 m^2. The lift of the aircraft causes downward motion of the double vortex structure at about $2.3 \pm 0.2\ m\,s^{-1}$. The theory of the Crow (1970) instability predicts a wavelength of the most unstable mode of approximately 400 m. The distance between the cores of the two trailing vortices (for an elliptical wing) amounts to about 46 m. However, very little data are available to verify these estimates.

In order to estimate the vertical motion and lateral dimensions of the vortex structure behind an aircraft, Baumann *et al.* (1993) measured the structure of contrails during the vortex regime as produced by a civil airliner. The measurements have been made by remote sensing from the Falcon research aircraft of the DLR flying about 1200 m above and behind civil airliners at cruise altitude. Due to its lower speed, the horizontal distance between the observed aircraft and the Falcon increases with time. The actual distance has been estimated from radar

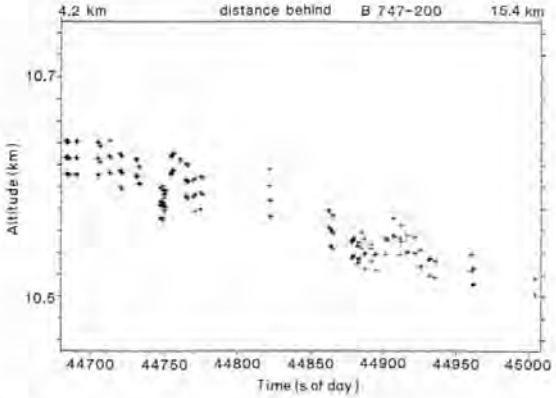

Fig. 15. Altitude of contrail of a B747-200 versus distance (approximately proportional to time of measurement)

observations and the time of measurement. The contrail is observed from above by a lidar system and by a video camera. The lidar emits laser light pulses, determines the distance of the backscattering contrails and the amount of light backscattered from the particles within the contrails. Using data on the actual position of the Falcon, these lidar signals are used to determine the altitude and lateral structure of the contrails.

Figure 15 shows the measured altitude of backscattering parts of contrail versus the distance behind the airliner. In this case the contrail was induced by a B747-200, flying at 10.7 km altitude and a true airspeed of $225\ m\,s^{-1}$ over southern Germany (49°N, 12°E), on 12:24:40–12:30:08 UTC, 9 April 1991, heading 310°. The contrail

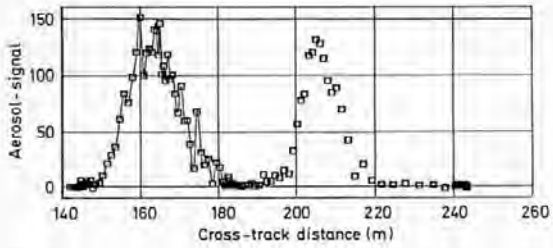

Fig. 16. Lateral structure of the contrail at 4.4 km behind a B747-200, as indicated by the integrated backscatter signal per laser shot versus cross-track position in m

Table 6. Measured scales of a contrail of B747-200

Distance behind aircraft (km)	Plume age (s)	Lateral distance (m)	Diameter of left contrail (m)	Diameter of right contrail (m)
4.4	19.6	42	14	11.5
4.8	21.3	44	12.5	11
5.2	23.1	41	15.5	14
5.7	25.3	44	12	11
7.0	31.1	36	10	9
7.2	32.0	51	10	14

was clearly visible for a distance of more than 15 km behind the aircraft. At a distance of about 7 km, the contrail becomes unstable, showing oscillating motions. These observations are documented by a video in addition to the lidar data. Figure 15 shows that the contrail sinks at an average speed of $2.4 \pm 0.6\ m\,s^{-1}$, amounting to about 120 m over a distance of 11.2 km, in ideal agreement with the above-mentioned theoretical estimate. Virtually the same values (120 m over 12 km distance) have been observed for the contrail of a DC-10.

Figure 16 shows the integrated backscatter signal as a function of the cross-track coordinate, 4.4 km behind the B747-200. The lateral distance between the two vortex cores of the contrail is indicated by the distance between the two backscatter maxima. It amounts to 42 m in this figure. The diameter of each of the two vortex cores forming the contrail is defined by the distance between the positions where the backscatter signal reaches 50% of the maximum value, giving 14 m (left) and 11.5 m (right) for this case. Table 6 lists the results for various distances behind the airliner. Again the measurements are in close agreement with the analytical estimate of 46 m. Table 6 shows that the lateral distance between the vortex cores is close to constant until about 5.7 km behind the B747, but then starts to oscillate at a wavelength of about 400 m, as expected for the Crow instability.

Table 7 presents mean concentrations in the wake in the young vortex regime. The computation assumes that the airliner consumes 60 t fuel per 5000 km or 12 g/m and that the emission indices are as given in Table 1. The resultant emissions are assumed to be evenly distributed over $5000\ m^2$, as estimated above for the end of the jet

Table 7. Mean concentration increases in trail after jet airliner

Emission product	Concentration increase	Background concentration at 8 km
CO_2	14 ppmv	354 ppmv
H_2O	14 ppmv	20–400 ppmv
NO, (NO_2)	78 ppbv	0.01–0.1 ppbv
CO_2	17 ppbv	40–100 ppbv
SO_2	3.1 ppbv	0.03–0.15 ppbv

regime behind a B747. The density of the environmental air is taken as $0.35\ kg/m^3$. The table gives the resultant concentration increases and provides typical orders of magnitude of environmental data for comparison. All the listed concentration changes are of measurable magnitude. Again we expect the largest effect with respect to NO_x and SO_2. The change in water vapour concentration is rather small and does not explain why large contrails are observed rather often.

Arnold *et al.* (1992) performed measurements using the DLR research aircraft Falcon to fly into several trails of commercial aircraft at flight altitude. The dusty plume was clearly visible and the wake turbulence of the airliners was still very notable, so that one was sure to measure inside the plume. This is corroborated by high-resolution humidity and temperature signals. Using a novel aircraft-borne automatic mass spectrometer (AAMAS), the abundances of the odd-nitrogen gases NO, NO_2, HNO_2, and HNO_3 were measured in the young trail (trail age $t_p = 9$ s corresponding to a distance of about 2 km) of a DC-9 airliner at 9.5 km altitude. AAMAS simultaneously measured SO_2 and H_2SO_4.

Figure 17 shows an example of the result from a measurement of the odd-nitrogen abundances. The HNO_2 abundances increased from about a background value of 5 pptv to a maximum of 520 pptv. Nitric acid is closely correlated with nitrous acid and reaches a maximum abundance of 460 pptv in the trail which exceeds the background abundance (about 40 pptv) by a factor of ten. Within the exhaust plume of the DC-9, the figure shows high abundances of NO and NO_2. Arnold (personal communication, 1993) recently found that the absolute values of measured NO and NO_2 need to be reduced by about a factor of ten. However, the data show at this distance that more than 80% of the NO_x emissions are composed by NO, and less than 1% of the NO_x emissions have the photochemically less reactive form of HNO_3. Also, the rather low values would hardly favour the formation of NAT particles. However, as explained by Fig. 18, Arnold *et al.* (1992) point out that the emitted water vapour causes supersaturated conditions in the plume of an aircraft and hence may initiate NAT particle formations. The formations of NAT particles may contribute to chlorine activation and ozone destruction.

Measurements of SO_2 have been performed in the same wake and in several further cases. Figure 19 shows, for example, the results obtained behind a B757 aircraft at a distance of 17.8 km and at an altitude of 11.3 km. The concentration reaches a maximum value of about 1 ppbv.

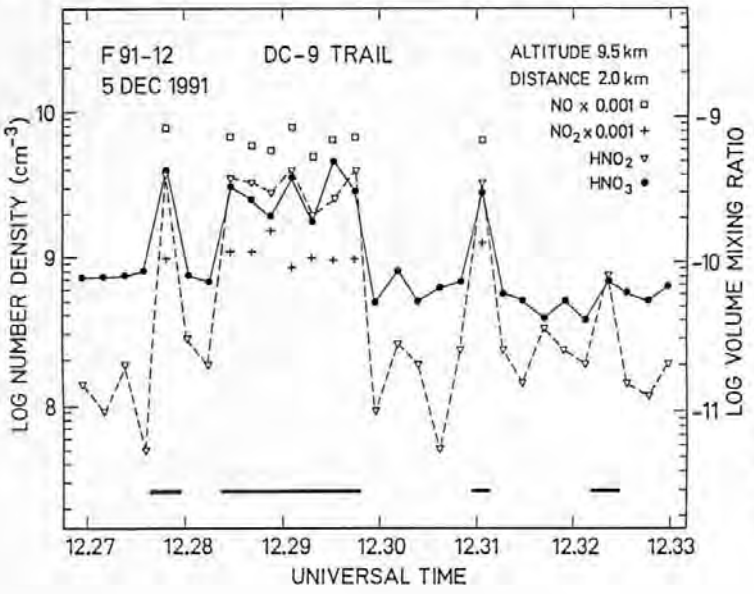

Fig. 17. Time plot of nitrous acid (HNO_2) and nitric acid (HNO_3) abundances measured during chase of a DC-9 airliner at 9.5 km altitude and a distance of 2 km. Periods when the research aircraft was inside the exhaust-trail of the DC-9 are marked by *bars*. For these periods NO and NO_2 abundances are also given, see text (from Arnold *et al.*, 1992)

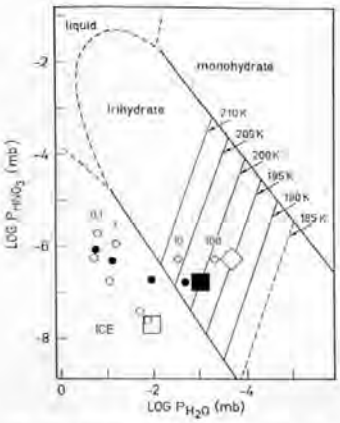

Fig. 18. Phase diagram of the HNO_3–H_2O system as obtained from laboratory measurements of Hanson and Mauersberger (1988), with symbols indicating typical conditions in the aircraft plume at various plume ages (from 0.1 to 100 s) (small symbols) in and in the environment (large symbols), at various altitudes: ◇: stratosphere at 20 km, ●: lower stratosphere, ○: upper troposphere. The water vapour concentration increases from right to left. The diagram shows that the atmosphere tends to form ice particles in the troposphere, but NAT particles (nitric acid trihydrate) in the lower stratosphere and even more in the middle stratosphere, provided the temperature is as low as indicated. Within the aged plumes, the NAT particles may form at higher temperatures than in the environment. For ambient temperatures which allow the existence of NAT particles, the plumes may provide the supersaturated conditions which are expected to be required for the initial NAT particle formations. Hence, PSC formation is more likely in the plume of stratospheric aircraft than in the undisturbed environment, mainly because of the higher water vapour concentration in the plume. From Arnold *et al.* (1992)

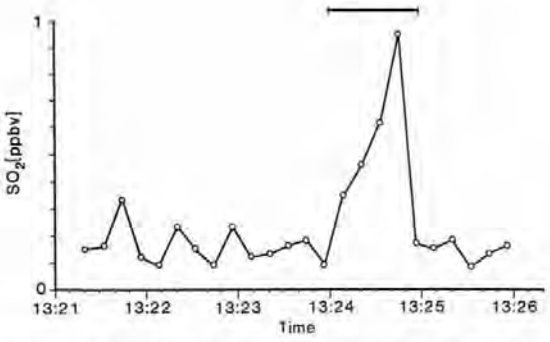

Fig. 19. SO_2 mixing ratio in the wake (trail) of as B757 aircraft

The measured background concentration of 100 pptv corresponds to previous findings (Georgii and Meixner, 1980). The value is smaller than estimated in Table 7, which is presumably caused by a larger area of mixing at this distance. Moreover, the measurements indicate that some portion (preliminary estimates give 1%) of the sulphur is converted in H_2SO_4. This is an important finding. It suggests that jet aircraft may form contrails composed of sulphuric acid aerosols. These aerosols do not evaporate after plume dissipation because the background atmosphere, around 10 km, is supersaturated with respect to liquid H_2SO_4–H_2O. The aerosol may provide CCN and sites for heterogeneous chemical reactions of trace gases, thereby possibly having an impact on the atmospheric environment.

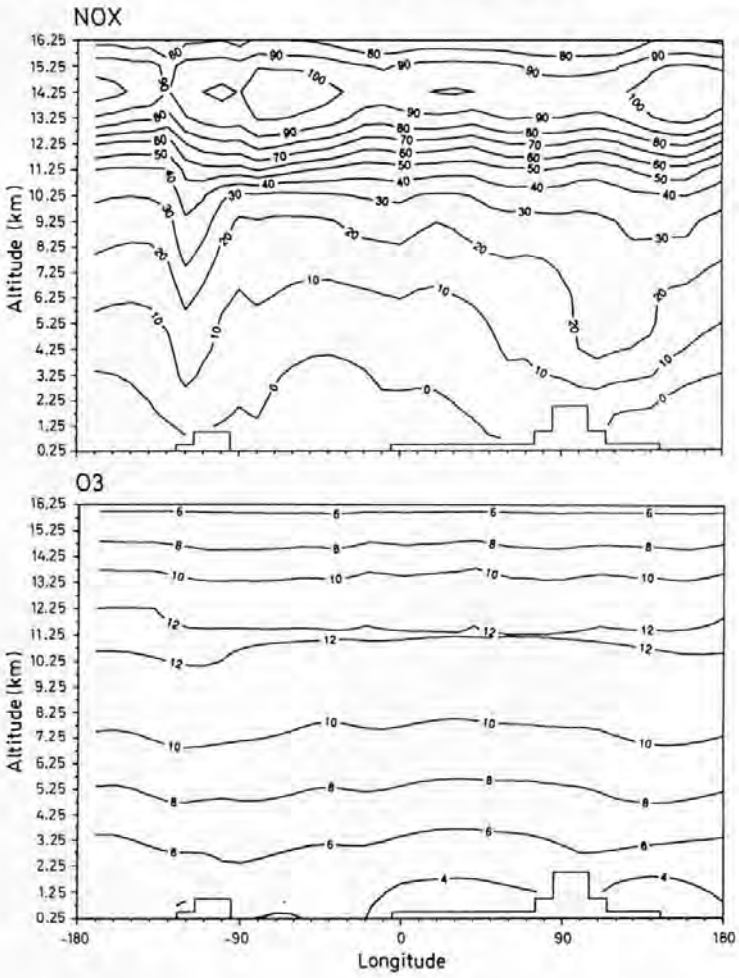

Fig. 20. The percentage change in the NO_x (top) and O_3 (bottom) concentration relative to a standard run without aircraft emissions due to the 1987 civil aircraft emissions, for April conditions, from Beck *et al.* (1992)

These are the first direct measurements of the chemical compositions in the wake of an airliner at cruise altitude. Douglass *et al.* (1991) presented measurements showing peaks in NO_2 and NO_y but were not sure that these peaks result from aircraft emissions.

7 Effects of emitted nitrogen oxide

Only a few studies of the effects of impact of aircraft emissions on air chemistry are known. Most studies concentrated on the effects from high-speed air traffic in the stratosphere (e.g. Johnston *et al.*, 1989, Jackman *et al.*, 1991). Early exceptions are Isaksen (1980) and Derwent (1982) who investigated the potential effects of air traffic operations in the troposphere using a 2D (latitude–altitude) transport-kinetics model. Their investigations were recently refined by Johnson *et al.* (1992). Similar investigations have been performed by Wuebbles and Kinnison (1990), Crutzen and Brühl (1990), Beck *et al.* (1992) and the WMO (1992). Up to the present, all studies neglected the impact of heterogeneous chemistry for subsonic air traffic. For illustration, we include Fig. 20 as published in Beck *et al.* (1992). The computations show that aviation emissions cause an increase of NO_x surpassing 100% near the tropopause. This supports the above corridor estimate in Table 5. The resultant ozone changes are approximately 12% slightly below the tropopause and positive everywhere.

These studies are not quite consistent because of different models and different emission rates. However, the studies agree in the general conclusion that the present rate of emissions from aircraft causes a notable increase in ozone in the upper troposphere: 4% (Derwent, 1982), 7% (Johnson and Henshaw, 1991), 12% (Beck *et al.*, 1992) and 15% (Isaksen, 1980). Significant changes to ozone occur in regions with low background values of nitrogen oxide (WMO, 1992). Johnson *et al.* (1992) find the largest changes in ozone concentration to occur in the southern hemisphere. It is clear that NO_x emitted from aircraft in

the upper troposphere is much more effective in enhancing ozone than ground-based emissions of NO_x (a factor of 17 in the calculations of WMO, 1992). For the future, using various assumptions on extrapolated emissions, the ozone change may reach 20–30% locally in the troposphere (Beck *et al.*, 1992). Following Liu *et al.* (1980), Beck *et al.* (1992) suggest that most of the presently observed ozone increase (see Fig. 2) in the upper troposphere is likely to be caused by NO_x emissions from air traffic. The observed changes in the trends (see Fig. 3), with steady ozone concentration in the upper troposphere in recent years, may be caused by a saturation effect in that the ozone increase per unit amount of NO_x emission decreases when the background concentration of NO_x increases.

The other emissions have a much smaller effect on ozone in these calculations. Both CO and the non-methane hydrocarbons are less efficient ozone producers than methane (WMO, 1992). Beck *et al.* (1992) and Johnson and Henshaw (1991) show that the ozone changes are virtually independent of the HC and CO emissions. Hence, the uncertainty in their emission factors is of minor importance, at least with respect to global ozone chemistry.

The above-mentioned studies concentrated on the global impact of NO_x. The chemistry in the immediate wake of aircraft has been studied recently by Weibrink and Zellner (1993), Danilin *et al.* (1992), and Miake-Lye *et al.* (1993). Since most of the NO_x emissions have the form of NO, a rapid but local destruction of ozone is to be expected. The effects depend strongly on the rate of mixing of the exhaust plume with the environmental air. Danilin *et al.* (1992) also considered the heterogeneous reaction given in Eq. (1), but found that this reaction does not play an important role at a timescale of up to 1 h in the wake. But this reaction can increase the HNO_3 in a plume near the tropopause by a factor of approximately two per week, causing a reduced effectivity of the ozone depletion potential.

With respect to the global warming effect from aircraft NO_x emissions, it is found that their specific effect is considerably larger (about 30-fold according to Johnson *et al.*, 1992) than for man-made emissions of NO_x at the surface. This is due to the larger changes in ozone and the larger radiative effects of ozone near the tropopause. These differences occur because the tropopause is cold, so that greenhouse gases have the strongest effect there and because the lifetime of NO_x is rather large and the background concentration of NO_x is low at this altitude, so that the photochemical effect of added NO_x is large. Lacis *et al.* (1990) use a 1D radiative-convective equilibrium model and show that the surface temperature should warm in response to both decreases in ozone above 30 km and increases in ozone below 30 km. Observed ozone trends suggest a cooling of the surface temperature of 0.05 °C. However, ozone changes due to NO_x emissions from subsonic air traffic cause a heating effect. The absolute warming effect from present aircraft emissions is about 0.02 K (Johnson and Henshaw, 1991) a figure which is about consistent with the findings of Lacis *et al.* (1990) and might reach a value of 0.2 K by the year 2050 (Brühl and Crutzen, 1988). However, these results were obtained for fixed cloud amounts and without accounting for heterogeneous chemistry on sulphuric acid aerosols, PSCs or ice clouds.

The increase in tropospheric ozone may offset the decrease in stratospheric ozone due to release of chlorofluorocarbons (Derwent, 1982) and even reduce the solar UV-B radiation at the earth surface in the industrialized northern hemisphere, in particular in summer (Brühl and Crutzen, 1989).

8 Effects of emitted water vapour and cloud formation

Water vapour alone is one of the most important greenhouse gases. In a warmer world, an increase of water vapour is expected which may about triple the greenhouse effect of carbon dioxide. The greenhouse effect of added water vapour (or any other infrared absorber) is strongest when added near the tropopause, as shielding of radiation to space is most pronounced if the absorber is cold (Graßl, 1990; Lacis *et al.*, 1990). As an absorber of terrestrial radiation in the stratosphere, one water vapour molecule is up to 200 times more effective than a CO_2 molecule (Graßl *et al.*, 1991).

As shown by simple radiation transfer calculations (Schumann and Wendling, 1990), an increase in water vapour concentration from its standard value to saturation in (an arbitrarily selected) altitude range 9–14 km causes changes in the local heating rate of order 1 K/h which is about 100 times larger than the effect of doubling CO_2. Although such a drastic change in water vapour concentration will not occur globally, it nevertheless demonstrates the strong radiative sensitivity of the atmosphere with respect to water vapour concentrations. A similar computation has been reported by Shine and Sinha (1991), who find that the sensitivity of the surface temperature to a fixed absolute increase in water vapour mixing ratio in individual global 40-hPa slabs is largest when the increase occurs in a slab at altitudes with pressures between 400 and 100 hPa. In absolute terms, the increase in surface temperature is of the order of 0.02 K for an increase of 0.001 g per kg in such a 50 hPa-slab.

Tables 3 and 4 indicate that the global contribution of water vapour emissions is small compared to the background state. However, the emissions add to the expected increases in water vapour concentrations due to the warming of the troposphere and add to stratospheric water vapour from the increasing rate of methane oxidation.

Even larger changes occur if the water condenses and forms thin ice clouds (Liou, 1986). Such ice clouds have a strong greenhouse effect if their albedo is low but the emissivity is large, which is the case for thin clouds with large (about 0.3 μm in radius or larger) particle sizes (Stephens and Webster, 1981; Stephens *et al.*, 1990). Liou *et al.* (1990) computed from a 2D model an increase in surface temperature of 1 K for a 5-% increase of high cirrus clouds (increase of total coverage by high clouds by about 1%) between 20°N and 70°N. Recently, Strauß and Wendling (DLR, personal communication, 1992) applied Liou's model in a regionalized version to the specific

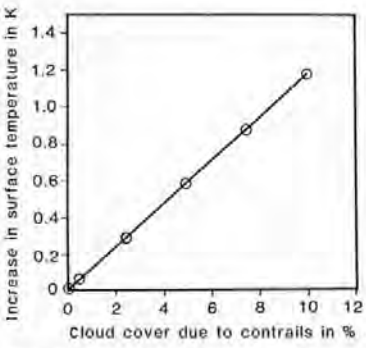

Fig. 21. Increase in surface temperature as a function of percentage cloud cover due to contrails. This is the result from a cloud radiation model which is adjusted to the local climatic conditions of South Germany

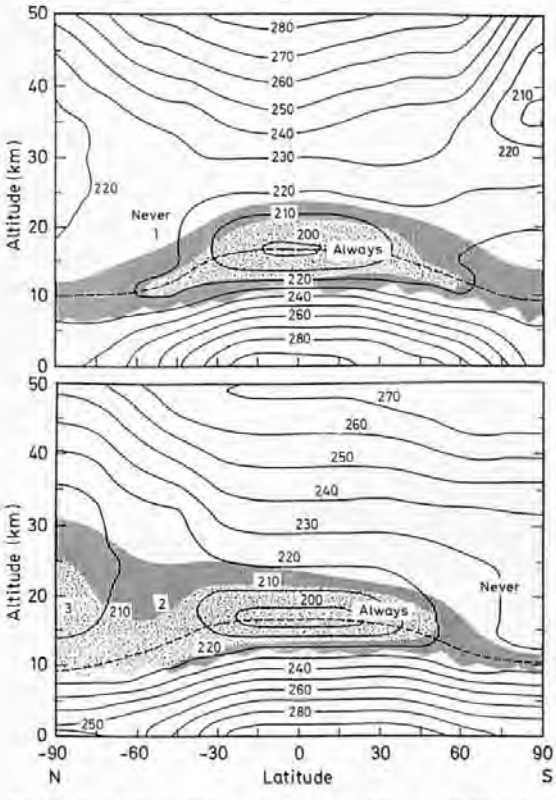

Fig. 22. Map versus latitude from pole to pole and altitude from surface to 50 km showing shaded regions where contrails from ALWAYS, eventually, or NEVER, assuming a standard atmosphere and contrails in local equilibrium after complete mixing. *Contours* represent constant temperature at increments of 10 K. Top: for 22 March, bottom: for 15 January. From Miake-Lye *et al.* (1993)

climatic conditions in southern Germany. They find that the surface temperature increases almost linearly with cloud cover due to contrails (see Fig. 21). For 0.4% additional cloud coverage by contrails, which is the presently observed value (Schumann and Wendling, 1990), the model predicts an increase in surface temperature by about 0.05 K. The regional effect is smaller because some of the feedback effects used by Liou (e.g. polar albedo change) are unimportant in this regional analysis.

Contrails emerge in particular at temperatures below $-40\,^{\circ}C$ (Appleman, 1953). Such low temperatures prevail in the tropopause, where temperatures between $-40\,^{\circ}$ and $-70\,^{\circ}C$ are typical for extratropical regions. Miake-Lye *et al.* (1993) have applied the analysis of Appleman (1953) to the standard atmosphere as a function of altitude and latitude. The result (see Fig. 22) shows regions labelled with ALWAYS corresponding to regions with temperature low enough for contrails to form even in dry air. The region labelled NEVER corresponds to warmer parts where contrails would not form even in a saturated atmosphere. By inspection we see that much of the high-flying air traffic takes place at altitudes where the formation of contrails is very likely, in particular in the northern winter hemisphere. A small reduction of the global mean temperature near and above the tropopause by, say, 2 K would greatly expand the region in which contrails are to be expected. Note that this analysis only considers the formation process of contrails. The residence time of such contrails depends strongly on the relative humidity of the environmental air and the amount of mixing with the contrail. The relative humidity is lower in the stratosphere but the mixing will be more rapid in the troposphere. Since very long living contrails will occur only at high relative humidity, the upper troposphere is more likely covered by large amounts of cirrus clouds induced by aircraft than the lower stratosphere.

Contrail observations from satellite data, lidar measurements, and climatological observations of cloud cover changes have been described in Schumann and Wendling (1990). Large (1–10 km wide and more than 100 km long) contrails are observed regionally on about 25% of all days within 1 year, but the average contrail coverage is only about 0.4% in central Europe.

Figure 23 shows a further example of lidar observation of contrails formed by air traffic. The lidar observations have been made on board of the Falcon aircraft at an altitude of about 12 km looking downward and passing over the airway G104 in southern Germany on 7 November 1990, 15:20 local time. The plot shows the backscatter signal which indicates a rather young contrail on the right and two older contrails on the left side. The thickness of the contrails is approximately 1.5 km with a core of 300 m. The width shown reaches 10 km but is actually smaller by a factor of 0.64 because the Falcon crossed the airway at an angle of about 40°. The core of the contrail with strong backscatter signals indicates high particle concentrations. Below that core we see the trace of sedimented particles with motions which are obviously affected by wind shear. Note that such sedimentation may contribute to a dehydration of the upper troposphere, possibly reducing the greenhouse effect.

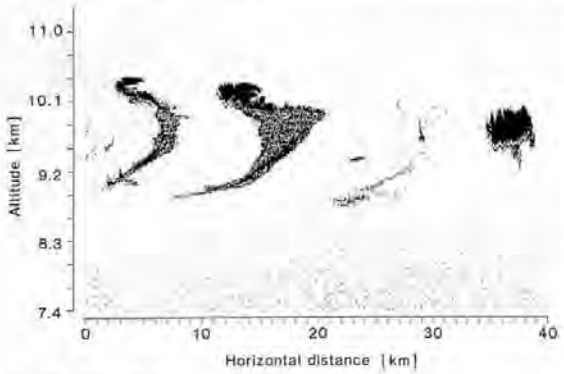

Fig. 23. Vertical cross section with contrails. The *grey tones* represent the backscatter signal amplitude as obtained from a Lidar sounding on board of the Falcon (flying from left to right) at 12 km altitude on 7 November 1990, 14.20 UTC

The lidar observations show that the contrails may become more than 1 km thick. Such big contrails cannot be explained by the amount of water which is emitted by the aircraft. Obviously, the large contrails form by triggering ice formation in the air which is subsaturated with respect to liquid water but supersaturated with respect to the ice phase (Detwiler and Pratt, 1984). The emitted sulphuric particles may also trigger such ice clouds in supersaturated regions. Hence, the largest artificial contrails are to be expected in regions with low ice nuclei concentration and where the water vapour concentration is above saturation with respect to the ice phase but below the saturation with respect to water. Unfortunately, the present data on water vapour concentrations from the global operational observation system are too inaccurate to determine these regions.

9 Summary

This survey reveals that high-flying subsonic air traffic affects the present state of the atmosphere mainly by its emissions of NO_x at high altitudes. As shown by simple estimates of mean concentration changes, NO_x may have doubled the background concentration in the upper troposphere at northern mid-latitudes, in particular in the flight corridors over the north Atlantic. Such changes may have caused about 5–20% change for ozone in the upper troposphere. It appears possible that the current air traffic is responsible for part of the observed ozone increase in the upper troposphere (Beck *et al.*, 1992). However, the increase of tropospheric ozone at flight altitudes has decreased in recent years which may signal a saturation effect of ozone increase because of increasing pollution. The greenhouse effect of the present change in ozone is estimated to be 0.01–0.02 K (Johnson *et al.*, 1992).

With respect to additional cloud cover, the estimates are very uncertain. Regional changes between a fraction of a percent and 2% appear possible. Analyses of satellite data for 1 year over Central Europe indicate 0.4% additional cirrus clouds from contrails. Lidar observations show that very large contrails may be formed with small particles which grow and sediment quickly. Hence, the additional cloudiness induced by contrails can be much larger than that to be expected from the added water vapour (which is small) because of ice nucleation triggered by the contrails in air supersaturated with respect to ice. Very thin ice clouds cause warming while thicker clouds cause cooling. Sedimentation of large ice particles may dehydrate the upper troposphere and, hence, reduce the warming effect. It is uncertain at the moment whether the warming dominates but this is what models predict. Quantitatively, the computed warming effect at the surface is not very large (0.01–0.05 K), but it is not clear whether this is the most meaningful impact indicator.

With respect to SO_2, the results suggest that air traffic increases the aerosol concentration in the lower stratosphere and upper troposphere. The aerosol may provide cloud condensation nuclei and sites for heterogeneous chemical reactions of trace gases, thereby possibly having an impact on the atmospheric environment. The increased concentrations of water vapour may also increase the amount of PSCs with possible negative effects on the ozone layer. However, these effects have not yet been quantified.

For the future, the strong increase in air traffic will be accompanied by a weaker but nonetheless important increase of about 2–4% in fuel consumption per year, meaning a doubling within the next 20 years or so. The climate changes induced by these emissions enhance the problems from other sources (e.g. reduced stratospheric temperature due to CO_2 increases or increased water vapour concentration due to increased methane concentrations and related increase in PSCs and reduced ozone).

Many questions have been identified which require further research. The most important ones are: what are the effective emission rates from present and future traffic at all levels of the atmosphere? How quick is the 3D dispersion and conversion from jet exit and flight corridors to global distribution? What are the chemical conversions, ozone generation and heterogeneous reactions on PSCs, ice crystals and aerosols? How large is the global area in which the water vapour concentration is above saturation with respect to the ice phase but below the saturation with respect to water, i.e. where large contrails may be formed? What are the climatic and environmental effects of the atmospheric changes for the living conditions on earth?

Corresponding 3D global circulation models including a sufficiently complete chemical cycle and radiation scheme for both the troposphere and the stratosphere are still under development (Dameris, 1993). More basic work is required to provide suitable models for the heterogeneous reactions. Jones *et al.* (1993) show that there is also considerable variability between models in the calculated photolysis rates of HNO_3, which increases the uncertainties of the computed effects of nitrogen oxide emissions on ozone. The 3D models are extremely computer time consuming because of the large number of chemical components and reactions to be treated. Moreover, the models have to provide sufficient resolution, in particular near the tropopause. Model validation studies are needed,

especially to verify the correct treatment of the interaction between stratosphere and troposphere. Also, the models need reliable estimates on natural emissions and background concentrations as input, which are not readily available. Of utmost importance are observational programmes to see whether the essential effects are included in the models and data to verify the model predictions. The North Atlantic flight corridor appears to be particularly suited for such validation measurements, because the impact of air traffic emissions should be of measurable magnitude in this region and clearly distinguishable from other causes.

Certainly, the list of uncertainties is long. Some of the effects of emissions on environment and climate may be small or even may mitigate the impact of other, more important disturbances to ozone layer and greenhouse effects. Nevertheless, the potential of aviation emissions to cause serious changes to climate and ozone layer exists. Limitations of the emissions from air traffic may become necessary sooner or later. Therefore, efficient means have to be developed to reduce the specific emissions from air traffic, in particular with respect to NO_x but also with respect to H_2O and SO_2.

Acknowledgements. I am grateful to F. Arnold (Max-Planck-Institut für Kernphysik, Heidelberg), D. Ehhalt and F. Rohrer (Forschungszentrum Jülich), B. Fay (Deutscher Wetterdienst Offenbach), Lufthansa Frankfurt, R. C. Miake-Lye (Aerodyne Research Inc., USA), K. Wege (DWD Hohenpeißenberg) and to several of my colleagues in DLR for providing unpublished research results and support. I thank A. Girard and N. Louisnard of ONERA for critical remarks which helped me to improve the paper. The author also gratefully acknowledges the support by the Deutsche Forschungsgemeinschaft (DFG), the Bundesministerium für Forschung und Technologie (BMFT), and the Commission of the European Community. This paper is a revised version of a key note paper at the AGARD Propulsion and Energetics Panel 81st Symposium held in Fiuggi, Italy, 10–14 May 1993.

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